Supplementary Materials for

# Incommensurate spin correlations in highly oxidized cobaltates $La_{2-x}Sr_xCoO_4$

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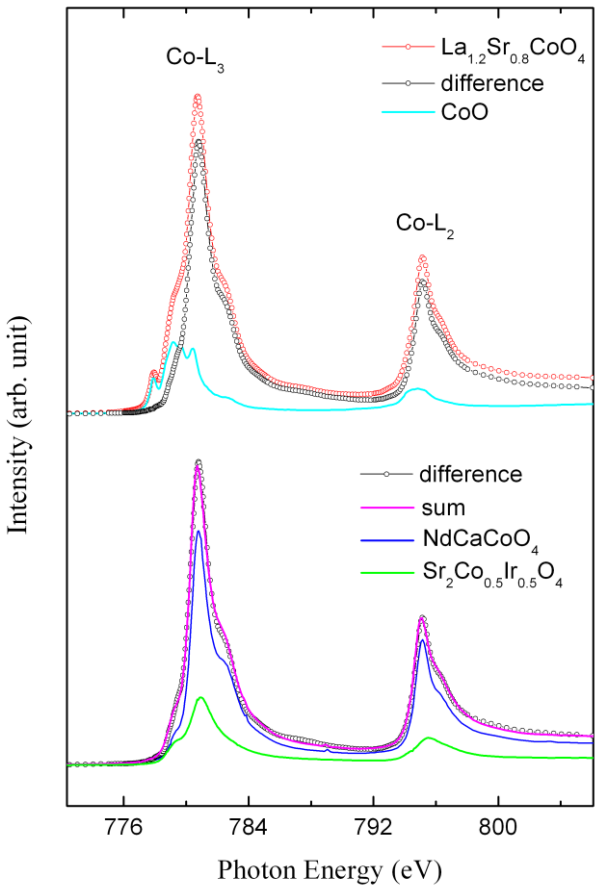


**Fig. S1** The Co-$L_{2,3}$ XAS spectrum of $La_{1.2}Sr_{0.8}CoO_4$ is shown together with XAS spectra of CoO, $NdCaCoO_4$ and $Sr_2Co_{1/2}Ir_{1/2}O_4$ reference samples. The pre-peak at 778 eV can be attributed to the presence of a $Co^{2+}$ species. The experimental Co-$L_{2,3}$ spectrum of $La_{1.2}Sr_{0.8}CoO_4$ can be decomposed into 77% $Co^{3+}$ content, denoted as 'difference' (black circles) and 23% $Co^{2+}$ contribution (cyan line). These measurements show that our $La_{1.2}Sr_{0.8}CoO_{4+\delta}$ sample has only a tiny oxygen deficiency ($\delta$=-0.015±0.005). After subtraction of the $Co^{2+}$ contribution (denoted as 'difference'), we were able to estimated the HS content for the $Co^{3+}$ species. Therefore, we used $NdCaCoO_4$ and $Sr_2Co_{0.5}Ir_{0.5}O_4$ [S1,S2] as a $Co^{3+}$ LS and $Co^{3+}$ HS reference material, respectively. The lower part of this figure shows that the difference spectrum can be nicely reproduced by the weighted sum (magenta) of the $Co^{3+}$ HS spectrum (green line) and the $Co^{3+}$ LS spectrum (blue line). Thus, the presence of ~19% $Co^{3+}$ ions in the HS state and ~58% $Co^{3+}$ ions in the LS state is revealed by this modelling of the 'difference' spectrum (besides 23% $Co^{2+}$ ions).

**References**

[S1] X. Ou and Hua Wu, Phys. Rev. B **89**, 035138 (2014)
[S2] unpulished results

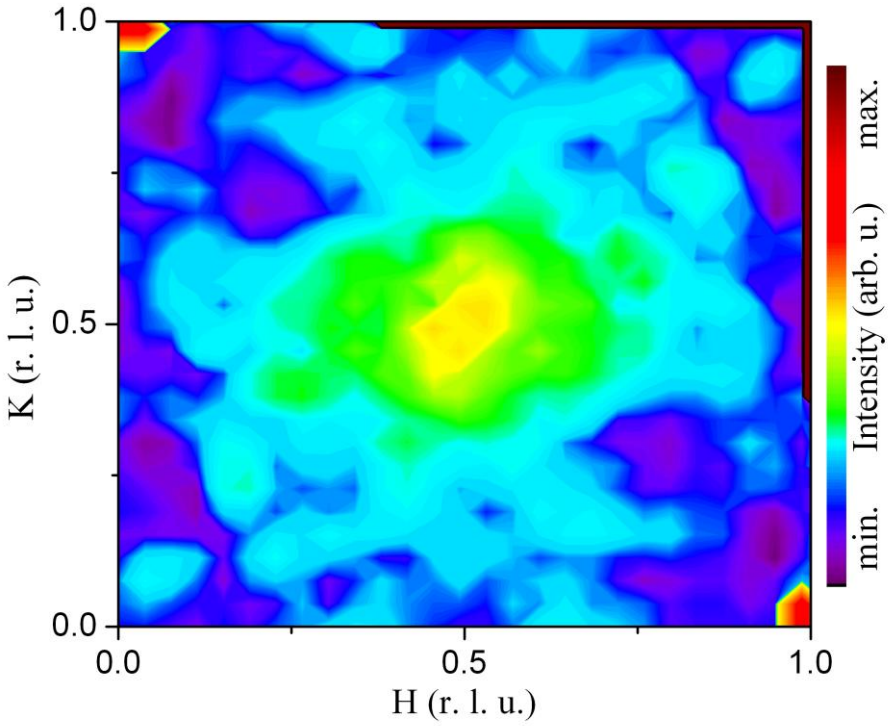


**Fig. S2** Fourrier transformation of charge ordering pattern used for numerical simulations in Fig. 6 (a). The expected intensity for charge correlations is shown in a logarithmic colour-contour plot. Since only the modulations of the Co-oxygen bond distances can be seen with neutrons and not the charges itself, the values are arbitrary.

# Supplementary Video Legends

## Supplementary Video #1: **Low energy magnetic excitations**

The movie shows a simulation of spin excitations at ~1.3 meV within our nano phase separation model for $La_{1.3}Sr_{0.7}CoO_4$.

## Supplementary Video #2: **Higher energy magnetic excitations**

The movie shows a simulation of spin excitations at ~8.4 meV within our nano phase separation model for $La_{1.3}Sr_{0.7}CoO_4$.